

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

216376US0PCT

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

10/018854

INTERNATIONAL APPLICATION NO  
PCT/JP00/04070INTERNATIONAL FILING DATE  
JUNE 22, 2000PRIORITY DATE CLAIMED  
JUNE 25, 1999 (Earliest)

## TITLE OF INVENTION

GAS GENERANT COMPOSITION

## APPLICANT(S) FOR DO/EO/US

Dairi KUBO, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
  4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
  5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
    - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
    - b. ☒ has been communicated by the International Bureau.
    - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
  6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
    - a. ☒ is attached hereto.
    - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
  7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
    - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
    - b. ☐ have been communicated by the International Bureau.
    - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
    - d. ☒ have not been made and will not be made.
  8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
  9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
  10. ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
  11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
  12. ☒ A copy of the International Search Report (PCT/ISA/210).

## Items 13 to 20 below concern document(s) or information included:

13. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☐ Certificate of Mailing by Express Mail
23. ☒ Other items or information:

Drawing (2 Sheets)  
Form PTO 1449  
PCT/IB/304  
PCT/IB/308

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

10/018856

INTERNATIONAL APPLICATION NO.

PCT/JP00/04070

ATTORNEY'S DOCKET NUMBER

216376US0PCT

24. The following fees are submitted:.

**BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5)) :**

- ☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$1040.00
- ☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$890.00
- ☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$740.00
- ☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$710.00
- ☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$100.00

**ENTER APPROPRIATE BASIC FEE AMOUNT =****CALCULATIONS PTO USE ONLY**

\$890.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).

\$0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	27 - 20 =	7	x \$18.00
Independent claims	1 - 3 =	0	x \$84.00

\$126.00

\$0.00

Multiple Dependent Claims (check if applicable). ☐

\$0.00

**TOTAL OF ABOVE CALCULATIONS =**

\$1,016.00

☒ Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.

\$0.00

**SUBTOTAL =**

\$1,016.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).

\$0.00

**TOTAL NATIONAL FEE =**

\$1,016.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). ☐

\$0.00

**TOTAL FEES ENCLOSED =**

\$1,016.00

Amount to be:

refunded

\$

charged

\$

- a. ☒ A check in the amount of \$1,016.00 to cover the above fees is enclosed.
- b. ☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 15-0030. A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

**Surinder Sachar**  
Registration No. 34,423

**22850**

SIGNATURE

**Norman F. Oblon**

NAME

**24,618**

REGISTRATION NUMBER

DATE

Dec. 26 2001

2/17

## Specification

Gas Generant Composition

## 5 Technical Field

The present invention relates to a gas generant composition suitable for use in a gas generator for a occupant restraint system, such as an airbag and a pretensioner, used to protect occupants in an automobile from a motor vehicle collision and the like. More particularly, the present invention  
10 relates to a gas generant composition for producing burning characteristics suitable as the gas generant.

## Background Art

An airbag system, which is one of occupant restraint systems, has been widely adopted in recent years to improve safety of occupants in an  
15 automobile. The airbag system operates on the principle that a gas generator is operated under control of signals from sensors detecting a collision, to inflate an airbag so as to cushion the shock of the occupants from the collision. The gas generator is required to have the capability of producing a required and sufficient amount of clean gas containing no  
20 harmful substance within a desired time.

In recent years, as the substitution of metallic compound azides that have been used hitherto, gas generants containing organic compounds including nitrogen as fuel components, combined with inorganic oxidizing agents, have been proposed. These known gas generants have the

advantages of generating a large amount of gas and of being less in danger in the manufacturing process. However, many of the gas generants using the organic compounds containing nitrogen as fuel have the heat of combustion as high as 2,500J/g or more and thus the gas generated becomes

5 high in temperature and pressure, so that there is a need for the gas generator to have a lot of coolants or cooling media. In addition to this, a slag or a by-product made in the burning of the gas generants is high in temperature and thus in flowability, so that there is a possibility that the slag may flow out of the gas generator and, if the worst happens, may scald

10 the occupants. Although any gas generators can be allowed to be improved by using a lot of coolants or cooling media, they are increased in size, going against the trend toward the size reduction and weight reduction of the gas generator.

To solve this problem, there were proposed several methods for

15 forming a slag that exhibits a high viscosity even in a high temperature by adding a slag forming agent, for improvement of the slag collecting efficiency. Among others, Japanese Laid-open (Unexamined) Patent Publication No. Hei 4(1992)-265292 discloses the method in which a low-temperature slag-forming agent as typified by silicon dioxide and a high-temperature

20 slag-temperature forming agent which produces a solid having a melting point close to or more than the combustion temperature are both added to improve the slag collecting efficiency. However, the slag forming agent itself is little contributive to the generation of gas and, with an increase of the amount of slag forming agent added, the amount of gas generated

decreases. In addition, an increase of the amount of slag forming agent added causes a decrease in combustion speed, thus making it difficult to adjust the combustion speed of the gas generants.

On the other hand, the gas generants arranged in the gas generator are formed into a specified shape, in order to produce a controlled combustion stabilization of the gas generants and a controlled burning behavior of the gas generated at the combustion. The combustion speed of the gas generants varies depending on the constituent ingredients of the gas generant composition and the particle diameter of the molded products of the gas generants. For the gas generant of slow combustion speed, the molded products of the gas generants are decreased in unit form or increased in total superficial area so that the gas can be generated rapidly within a short time. On the other hand, for the gas generant of rapid combustion speed, the molded products of the gas generants are increased in unit form or decreased in total superficial area so that a desired gas generating behavior can be produced.

The burning characteristics of the gas generator are almost always determined by the burning behavior of the gas generants used. The burning characteristics of the gas generator are generally evaluated, for example, by a curve plotted between a tank internal pressure obtained by operating the gas generator in a 60 liter tank and time. In recent years, the so-called depower technique of protecting the occupants from possible harm at the inflation of the airbag has attracted public attention. For this purpose, the gas generator is now being desired to have such burning

characteristics that in the 60 liter tank test of the gas generator, the gas generation speed increases slowly in the period of 10-20 milliseconds from the ignition and then increases sharply on and after 20 milliseconds. The gas generator having this burning characteristic controls the gas generation speed moderately at the initial stage of the combustion, to provide a more ideal occupant protecting performance. The burning behavior of the gas generants can be partly controlled by changing configuration or form of the molded products of the gas generants and calculating the amount of gas generated. The relationship between the form of the molded products of the gas generants and the amount of gas generated is an old public knowledge in the field of propellant. For example, a suitable form of the molded products of the gas generants can easily determined by making reference to Explosive Handbook at page 279 (issued from Kyoritsu Shuppan Co., Ltd., 1987), for example.

Another available methods have been proposed in which at least two gas generant compositions of different combustion speeds are combined in layers so that the burning behavior can be controlled in stages. For example, Japanese Laid-open (Unexamined) Patent Publication No. Hei 6(1994)-48880 discloses the control of the burning characteristics of the gas generator by using this method. Also, Japanese Laid-open (Unexamined) Patent Publications No. Hei 6(1994)-107108 and No. Hei 6(1994)-107109 disclose the gas generants each having a coat of an inert combustion delay on a part thereof. Both of them disclose that the gas generant compositions of different combustion speeds are combined in layers to form the molded

products of the gas generants. When these gas generants are ignited, they are allowed to start burning from a relatively flame retardant layer and thereby the gas generating speed at the initial stage of the 60 liter tank test is suppressed. These gas generants require more processes for the molding than usual, because the gas generant compositions of different combustion speeds are combined in layers. Also, since at least two gas generant compositions are required, the gas generants are manufactured at considerable cost.

Further, Japanese Laid-open (Unexamined) Patent Publications No. Hei 10(1998)-87390 and No. Hei 10(1998)-324588 disclose the molded products of the gas generants formed into a specified form. These disclose that the gas generants are formed into a specific form, such that as the combustion of the gas generants proceeds, their combustion superficial areas are narrowed at a slowest possible speed, or would rather be enlarged, whereby a desired combustion performance is tried to be produced.

However, the both forms of the molded products of the gas generants have a single-hole or multi-hole (porous) tubular form and thus have cavities therein, so that when the gas generants are filled in the gas generator, the filling density is decreased. In addition, since the forms of the gas generants are limited to some specific forms, it is difficult to vary the forms of the gas generants in corresponding to various forms of the gas generators.

It is the object of the present invention to provide a gas generant composition, suitable for use in a gas generator for an automobile occupant restraint system, such as an airbag and a pretensioner, which exhibits the

burning characteristics to produce a further ideal occupant protecting performance, without any complex production process being required for forming the molded products of the gas generants and without any limitation being imposed on the forms of the molded products of the gas generants.

### Disclosure of the Invention

The inventors have been devoted themselves to consider possible ways of solving the problems above and found that the burning behavior of the gas generant was improved by defining the composition of the gas generant and have derived the present invention therefrom.

A gas generant composition of the present invention is characterized in that it comprises a fuel, an oxidizing agent and an additive, wherein the fuel comprises at least one high-energy nitrogen-containing organic compound and at least one low-energy nitrogen-containing organic compound, and the low-energy nitrogen-containing organic compound has a 50% average particle diameter of  $40\mu\text{m}$  or less.

The combustion speed of the fuel depends heavily on the reactivity of the oxidizing agent used and the combustion speed. In the present invention, the importance of the combustion speed of the fuel was recognized for the sake of argument of the combustion speed, and a nitrogen-containing compound of high combustion speed was defined as a high-energy nitrogen-containing organic compound and a nitrogen-containing compound of low combustion speed was defined as a



low-energy nitrogen-containing organic compound.

Usually, when two or more different kinds of fuels are used as the fuel of the gas generant composition, the combustion speed does not exceed any combustion speed of the gas generant composition comprising single kind of fuel but stands at a nearly constant value of a generally intermediate level between their respective combustion speeds. However, when making a great difference between the energies the fuels have, the gas generant comprising such gas generant composition does not burn at a constant combustion speed. The combustion speed decreases at an initial stage in the form of the combustion of the high-energy fuel being inhibited by the low-energy fuel in a period from the ignition in the gas generator. As a result, the internal pressure of the gas generator fully increases and the combustion speed of the gas generant increases.

When the low-energy nitrogen-containing organic compound has the 50% average particle diameter of  $40\mu\text{m}$  or less, or preferably  $20\mu\text{m}$  or less, as in the present invention, the combustion inhibiting effect of the high-energy fuel by the low-energy fuel is produced moderately.

It is to be noted that the 50% average particle diameter of number of reference is a measurement by which a size distribution is expressed on the basis of number: when the total number of particles is taken as 100, the particle size obtained when the particles integrated from the smaller number reach 50 is called the 50% average particle diameter of number of reference.

In the 60 liter tank test, the gas generant comprising only the gas

generant composition of the present invention exhibits the burning behavior that it burns at a low combustion speed at the initial stage and then burns at an increased combustion speed. Thus, the gas generant comprising only the gas generant composition of the present invention burns at a varied  
5 speed, not at a generally constant speed. In contrast, the conventional gas generant comprising only one gas generant composition burns at a generally constant speed. Hence, the gas generant comprising only the gas generant composition of the present invention is fundamentally different from the conventional gas generant in this point.

10 Thus, the present invention can produce the desired burning behavior without any particular process, such as the process that two or more gas generant compositions of different combustion speeds are combined in layers to form the molded products of the gas generants or the process that the molded products of the gas generants are limited to some specific form,  
15 being required for obtaining the gas generant suitable for the gas generator for the automobile occupant restraint system such as the airbag.

In general, the gas generant composition is selected from the gas generant compositions whose combustion speed can be given approximation to a generally constant combustion speed in the actual operation pressure  
20 range of the gas generator. This is because, if a gas generant composition whose combustion speed overly depends on pressure is selected, the combustion speed is varied considerably by an environmental temperature change or a pressure change in the gas generator caused by degradation of the gas generants and the like, so that such a gas generant composition is

not desirable as the gas generant composition used for the gas generator for the automobile occupant restraint system such as the airbag.

The dependency of the combustion speed on the pressure is obtained by a pressure exponent of the following general formula on the combustion speed of explosives. The burning behavior originating from the form of the gas generant can be determined by primarily approximating the gas generating speed, from which the combustion performance of the gas generator can be approximately estimated:

$$V=aP^n \quad (\text{Vieille formula})$$

where "V" is a combustion speed; "a" is an exponent dependent on the composition and temperature; "P" is a pressure; and "n" is a pressure exponent.

It is preferable that a gas generant composition of relative low in pressure exponent and small in variation of combustion speed by the pressure in the general formula above has little dependency of the combustion speed on the pressure and thus is suitable as the gas generant composition used for the gas generator for the automobile riser restraint system such as the airbag.

The gas generant composition of the present invention exhibits the pressure exponent substantially equal to that of the conventional gas generant composition.

#### Brief Description of the Drawings

FIG. 1 is a schematic view of a gas generator 1 for an airbag used in respective embodiments of the present invention;

FIG. 2 is a graph showing the 60 liter tank test results; and

FIG. 3 is a table showing the 60 liter tank test results.

### Best Mode for Carrying out the Invention

A composition of a gas generant of the present invention comprises a  
5 fuel, an oxidizing agent and an additive. The fuel comprises at least one  
nitrogen-containing organic compound combustible at a high combustion  
speed and at least one nitrogen-containing organic compound combustible at  
a low combustion speed.

In other words, the fuel of the gas generant composition of the present  
10 invention comprises at least one high-energy nitrogen-containing organic  
compound and at least one low-energy nitrogen-containing organic  
compound. Further, the low-energy nitrogen-containing organic compound  
has a 50% average particle diameter of  $40\mu\text{m}$  or less, or preferably  $20\mu\text{m}$   
or less.

15 The gas generator using the gas generants comprising the gas  
generant composition of the present invention has such burning  
characteristics that in the combustion curve obtained by the 60 liter tank  
test, the combustion speed increases slowly in the period of before 20ms  
from the ignition and then increases sharply on and after 20ms. The gas  
20 generant composition of the present invention exhibits the pressure  
exponent of substantially the same level as that of the conventional gas  
generant composition. Thus, the use of the gas generant composition of the  
present invention to the gas generator can produce an ideal burning  
characteristic.

A further detailed description on the gas generant composition of the present invention will be given below.

The high-energy nitrogen-containing organic compound used in the present invention is the compound which is high in enthalpy of formation, relatively easy to combust and exhibits a rapid combustion speed. The high-energy nitrogen-containing organic compounds that may be used include those having the enthalpy of formation of  $-200\text{kJ/mol}$  or more (standard condition), or preferably  $-100\text{kJ/mol}$  or more. Specifically, at least one high-energy nitrogen-containing organic compound selected from the group consisting of aminotetrazole, nitroguanidine, and triaminoguanidine nitrate can be cited as the concrete example of the high-energy nitrogen-containing organic compound.

The low-energy nitrogen-containing organic compound used in the present invention is the compound which is low in enthalpy of formation, difficult to ignite and exhibits a slow combustion speed. The low-energy nitrogen-containing organic compounds that may be used include those having the enthalpy of formation of  $-200\text{kJ/mol}$  or less (standard condition), or preferably  $-300\text{kJ/mol}$  or more. Specifically, guanidine nitrate and oxamide can be cited as the concrete example as the low-energy nitrogen-containing organic compound.

Although the present invention puts no limitation on the combination of the high-energy nitrogen-containing organic compound and the low-energy nitrogen-containing organic compound, the difference in enthalpy of formation therebetween is preferably  $200\text{kJ/mol}$  or more.

Aminotetrazole is of preferable as the high-energy nitrogen-containing organic compound in that it contains a high proportion of nitrogen and is relatively high in safety in the handling. Nitroguanidine is also preferable as the high-energy nitrogen-containing organic compound in that it is large  
5 in number of moles of the generated gas.

Guanidine nitrate is preferable as the low-energy nitrogen-containing organic compound in that it is relatively available and low in cost, though it is seldom used for the gas generator in that it usually burns at a slow combustion speed when singularly combined with oxidizing agent.

10 When aminotetrazole is used as the high-energy nitrogen-containing organic compound and guanidine nitrate is used as the low-energy nitrogen-containing organic compound, the difference in enthalpy of formation is 598kJ/mol. When nitroguanidine is used as the high-energy nitrogen-containing organic compound and also guanidine nitrate is used as  
15 the low-energy nitrogen-containing organic compound, the difference in enthalpy of formation is 296kJ/mol.

Mixing ratios between the high-energy nitrogen-containing organic compound and the low-energy nitrogen-containing organic compound are between 10:1 and 1:10, or preferably between 5:1 and 1:5 in a ratio of weight.

20 The content of the high-energy nitrogen-containing organic compound and the low-energy nitrogen-containing organic compound as the fuel to the total gas generant composition is 15-85 weight%.

Further, when aminotetrazole is used as the high-energy nitrogen-containing organic compound and guanidine nitrate is used as the

low-energy nitrogen-containing organic compound, the mixing ratios therebetween are preferably between 3:1 and 1:3 in the ratio of weight. When nitroguanidine is used as the high-energy nitrogen-containing organic compound and guanidine nitrate is used as the low-energy

5 nitrogen-containing organic compound, the mixing ratios therebetween are preferably between 5:1 and 1:1 in the ratio of weight. When an amount of nitroguanidine is excessive, the combustion speed is significantly decreased. On the other hand, when an amount of nitroguanidine is too little, an adequate combustion performance is not obtained.

10 It is preferable that the low-energy nitrogen-containing organic compound has the 50% average particle diameter of  $40\mu\text{m}$  or less, or preferably  $20\mu\text{m}$  or less. In fact, as the low-energy nitrogen-containing organic compound decreases in 50% average particle diameter, the combustion speed decreases in the period of before 20ms from the ignition of  
15 the gas generant composition. It is noted that it is difficult to pulverize the low-energy nitrogen-containing organic compound, nitroguanidine, in particular, and there have been no reported cases of nitroguanidine being pulverized to  $40\mu\text{m}$  or less for use as the gas generant. With the 50% average particle diameter of not less than  $40\mu\text{m}$ , the effect of the  
20 low-energy fuel cannot be provided, so that a sufficient decrease in combustion speed is not provided at an early stage of the combustion.

Further, sufficient collapse strength is not obtained in the press molding of the gas generant composition. With the 50% average particle diameter of  $5\mu\text{m}$  or less, the pulverization undesirably requires a

considerable cost, though the effect of the invention can be provided. It should be noted further that since the guanidine nitrate pulverized to 40  $\mu$ m or less serves as an adhesive agent in the press mold of the gas generant composition of the present invention, the pellets of the gas generant composition produced can have high collapse strength (high hardness).

The oxidizing agents that may be used in the present invention include oxyacid salts, such as nitrate, halogen acid salt and chromate, oxides, and peroxides. Any oxidizing agents may be used, as long as they can oxidize the fuel comprising the high-energy nitrogen-containing organic compound and the low-energy nitrogen-containing organic compound. Preferably, at least one material selected from the group consisting of nitrate, perchlorate, chlorate as concerns alkali metals or alkali earth metals and basic copper nitrate is used as the oxidizing agent. In addition, at least one material selected from the group consisting of mixtures of phase stabilized ammonium nitrate or ammonium perchlorate and nitrate, perchlorate, chlorate as concerns alkali metals or alkali earth metals and basic copper nitrate may also preferably be used as the oxidizing agent. Strontium nitrate that is converted into a high-viscosity, slag-forming metal component by burning is further preferable.

The content of the oxidizing agent can be determined within the range of nearly equivalent within which the gas generant composition is completely burnt stoichiometrically. In the gas generant composition containing the oxidizing agent largely deviating from such an equivalent,



CO or NO<sub>x</sub> in the combustion gas are significantly increased. The content of the oxidizing agent to the total gas generant composition can be determined from the range of 30 weight% to 70 weight%. With the content of the oxidizing agent of less than 30 weight%, there is the possibility that oxygen supply may fall short so that imperfect combustion may be caused to produce a harmful CO gas. On the other hand, with the content of the oxidizing agent in excess of 70 weight%, there is the possibility that the required gas for the inflation of the airbag may not be supplied.

Strontium nitrate, which is preferably used as the oxidizing agent in the present invention, can be used by itself or in the form of a mixed oxidizing agent by combining with alkali metal nitrate, ammonium perchlorate or basic copper nitrate. To give actual examples, combination of strontium nitrate and potassium nitrate, combination of strontium nitrate and ammonium perchlorate, or combination of strontium nitrate and basic copper nitrate can be cited.

The mixed oxidizing agent in the gas generant composition of the present invention can allow the combustion speed to increase, to provide a harmless combustion gas by adding a small amount of nitrate of the alkali metal and basic copper nitrate. Ammonium perchlorate is particularly suitable as the gas generant composition used for the pretensioner, because a small amount of ammonium perchlorate added can produce an increased number of moles of the generated gas by the gas generant.

When potassium nitrate is added, particular attention should be paid to the fact that the oxidizing agent can provide the effect with a low

potassium nitrate content of 10 weight% or less of the total amount of gas generant composition. With a potassium nitrate content of more than 10 weight%, the outflow slag generated by the combustion of the gas generant composition increases. It is hard to filter out the slag originating from the potassium through filters in the gas generator, so that there is the possibility that the slag may cause damage to the airbag or may scald the occupants. Also, if more potassium nitrate is used, it becomes hard to achieve the characteristic feature of the gas generant composition that the combustion speed is controlled moderately at the initial stage, so that there is provided an increased possible fear of doing harm to the occupants.

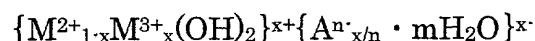
When basic copper nitrate is added, the content of basic copper nitrate is preferably 30 weight% or less of the total amount of gas generant composition. It is possible to filter out the slag originating from the basic copper nitrate with ease, differently from the use of potassium nitrate, so that a basic copper nitrate content of 30 weight% or less is permissible. If more basic copper nitrate is used, there is the possibility that the combustion speed of the gas generant composition may decrease so that a desired combustion speed cannot be obtained.

The additives that may be used include a slag forming agent and a binder. In the gas generant composition of the present invention, silicon nitride or silicon carbide is preferably used as the slag forming agent. While silicon nitride and silicon carbide, which are called fine ceramics, are used as heat-resistant materials which are thermally stable and high resistant, they have the property of being decomposed in high temperature

oxidizing atmospheres. Through the use of this property, both processes of the slag forming and the gas generation are provided. The content of silicon nitride or silicon carbide is preferably in the range of 0.5 to 10 weight%. With a less than 0.5 weight% silicon nitride or silicon carbide, the satisfactory effect cannot be expected in the slag collecting process. On the other hand, with a more than 10 weight% silicon nitride or silicon carbide, the content of fuel and oxidizing agent is decreased on the whole, so that there is the possibility of short amount of gas generated.

When fine particles of silicon nitride or silicon carbide are added in the pulverizing process of the fuel or the oxidizing agent, they provide the effect of acting as an anti-caking agent. Silicon nitride or silicon carbide has the property of providing the slag forming capability without decreasing the combustion speed of the gas generant composition containing the low-energy fuel. If a required amount of SiO<sub>2</sub> is added as the slag forming agent, the combustion speed is significantly decreased. Thus, the use of SiO<sub>2</sub> as the slag forming agent is not desirable for the present invention.

Further, hydrotalcites expressed by the following general formula are preferably used as the binder and slag forming agent:



where M<sup>2+</sup> represents bivalent metals including Mg<sup>2+</sup>, Mn<sup>2+</sup>, Fe<sup>2+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>, Cu<sup>2+</sup> and Zn<sup>2+</sup>;

M<sup>3+</sup> represents trivalent metals including Al<sup>3+</sup>, Fe<sup>3+</sup>, Cr<sup>3+</sup>, Co<sup>3+</sup> and In<sup>3+</sup>;

A<sup>n-</sup> represents n-valence anions including OH<sup>-</sup>, F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, CO<sub>3</sub><sup>2-</sup>,

SO<sub>4</sub><sup>2-</sup>, Fe(CN)<sub>6</sub><sup>3-</sup>, CH<sub>3</sub>COO<sup>-</sup>, ion oxalate, and ion salicylate; and

X:  $0 < x \leq 0.33$ .

Typical of the hydrotalcites are synthetic hydrotalcite expressed by the chemical formula of Mg<sub>6</sub>Al<sub>2</sub>(OH)<sub>16</sub>CO<sub>3</sub> · 4H<sub>2</sub>O or pyroaurite expressed by the chemical formula of Mg<sub>6</sub>Fe<sub>2</sub>(OH)<sub>16</sub>CO<sub>3</sub> · 4H<sub>2</sub>O. The hydrotalcites, which are a porous material having water of crystallization, are very useful as a binder for a gas generant of nitrogen-containing organic compound. The gas generant composition containing the hydrotalcite as the binder can provide a degree of hardness much higher than a degree of hardness of a pellet of a general type of azide base gas generant, particularly when used for the gas generant composition containing tetrazoles as the main component.

This seems to be because the hydrotalcites have the common property of being liable to absorb moisture, which property serves to firmly bind the components of the gas generant. Also, the pellets using this binder keep their own characteristic and flammability characteristic unchanged against the thermal shock caused by temperature being raised and fallen repeatedly, thus enabling the pellets to be minimized in deterioration with age after practical installation on a vehicle. Further, it is probable that for example, the synthetic hydrotalcite of the hydrotalcites is allowed to react as expressed by the following reaction formula (1) in the combustion of the gas generant.



Due to this, no harmful gas is produced. Also, the reaction itself is an

endothermic reaction, thus providing an advantageous effect of reducing the heat value of the gas generant. In addition, the decomposition product itself of the synthetic hydrotalcite forms spinel that can easily be filtered by the slag reaction of the acid base reaction expressed by the following  
5 formula (2).



The content of the binder to the total gas generant composition is preferably in the range of 2 to 10 weight%. With the binder content of less than 2 weight%, the function as the binder is achieved with difficulty. On  
10 the other hand, with the binder content of more than 10 weight%, the content of the fuel and oxidizing agent is decreased on the whole, so that there is the possibility of short amount of gas generated. Also, by adding the hydrotalcites to the gas generant composition, the gas generant composition is reduced in sensibility and, as a result of this, the effect of  
15 providing an improved safety in the production process is also yielded.

Also, cellulosic binders or natural polymers may be used as the binder. These binders are suitable for the extrusion molding of the gas generant composition. To give an actual example of the cellulosic binders, at least one material selected from the group consisting of carboxymethyl cellulose,  
20 methyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, and hydroxylpropyl methyl cellulose can be cited. To give an actual example of the natural polymers, at least one material selected from the group consisting of Cyamoposis Gum or tragacanth gum can be cited. The content of the cellulosic binder or natural polymer is preferably in the range

of 2 to 10 weight%.

To give an actual example of other binders than the cellulosic binders, at least one material selected from the group consisting of polyacrylic acid, sodium polyacrylate, polyacrylamide, and two or three copolymerized compounds can be cited. The amount of those binders added is preferably in the range of 0.5 to 10 weight%. The addition of this binder provides the effect of providing an improved heat resistance of the gas generant composition.

Addition of silane compounds can provide an improved moldability in the extrusion molding process, in particular. The silane compounds that may be used include, for example, vinyltrimethoxysilane, vinyltriethoxysilane, vinyltris( $\beta$ -methoxyethoxy)silane,  $\beta$ -(3,4-epoxycyclohexyl)ethyltrimethoxysilane,  $\gamma$ -glycidoxy propyltrimethoxysilane,  $\gamma$ -glycidoxy propylmethyl diethoxysilane,  $\gamma$ -methacryloxy propylmethyl dimethoxysilane,  $\gamma$ -methacryloxy propyltrimethoxysilane,  $\gamma$ -methacryloxy propylmethyl diethoxysilane,  $\gamma$ -methacryloxy propyltriethoxysilane, N- $\beta$ -(aminoethyl)- $\gamma$ -aminopropylmethyl dimethoxysilane, N- $\beta$ -(aminoethyl)- $\gamma$ -aminopropyltrimethoxysilane, N- $\beta$ -(aminoethyl)- $\gamma$ -aminopropyltriethoxysilane,  $\gamma$ -aminopropyltrimethoxysilane,  $\gamma$ -aminopropyltriethoxysilane, N-phenyl- $\gamma$ -aminopropyltrimethoxysilane, tetramethoxysilane, methyltrimethoxysilane, dimethyldimethoxysilane, phenyltrimethoxysilane, diphenyldimethoxysilane, tetraethoxysilane, methyltriethoxysilane, dimethyldiethoxysilane, phenyltriethoxysilane,

diphenyldiethoxysilane, hexyltrimethoxysilane, hexyltriethoxysilane, decyltrimethoxysilane, and hexamethyldisilazane. The amount of the silane compounds added is preferably in the range of 0.5 to 10 weight%.

Next, reference is given to a preferable combination of components of the gas generant composition of the present invention. A preferable combination of components of the gas generant composition is the combination of 5-aminotetrazole and guanidine nitrate used as the fuel, strontium nitrate used as the oxidizing agent, silicon nitride used as the slag forming agent, and synthetic hydrotalcite used as the binder. This gas generant composition preferably contains 10-30 weight% of 5-aminotetrazole, 5-30 weight% of guanidine nitrate, 30-70 weight% of strontium nitrate, 0.5-10 weight% of silicon nitride, and 2-10 weight% of synthetic hydrotalcite. Further, in order to produce an increased combustion speed, it is preferable that not more than 10 weight% of potassium nitrate or not more than 30 weight% of basic copper nitrate is contained. Guanidine nitrate is low in energy, so that it has the tendency to cause the combustion speed to decrease as the guanidine nitrate content increases. In addition, particles of the guanidine nitrate are so hard that they cannot be pulverized with ease. Although the particles of the guanidine nitrate of 50  $\mu$ m or more in 50% average particle diameter are easily obtained by use of a generally used pin mill or ball mill as pulverizing means used in the manufacture of the gas generant, since it is very difficult to pulverize the particles of the guanidine nitrate into 40  $\mu$ m or less, particularly 20  $\mu$ m or less, a special milling machine, such as a jet mill, must be used therefor. It is preferable

that silicon nitride is used as the slag forming agent. The addition of silicon nitride can provide good slag collecting properties without decreasing the combustion speed. Also, synthetic hydrotalcite should preferably be used as the binder in that it can provide not only improved hardness of the gas generant composition but also reduced heat release value and improved slag collecting capabilities.

Another preferable combination of components of the gas generant composition is the combination of nitroguanidine and guanidine used as the fuel, strontium nitrate used as the oxidizing agent, silicon nitride used as the slag forming agent, and a cellulosic binder used as the binder. This gas generant composition preferably contains 20-55 weight% of nitroguanidine, 5-30 weight% of guanidine nitrate, 30-60 weight% of strontium nitrate, 0.5-10 weight% of silicon nitride, and 2-10 weight% of cellulosic binder or synthetic hydrotalcite. Further, in order to produce an increased combustion speed, it is preferable that not more than 10 weight% of potassium nitrate or not more than 30 weight% of basic copper nitrate is contained. The gas generant composition of the present invention containing nitroguanidine and guanidine nitrate as the fuel is preferably produced in the form of the gas generant by extrusion molding. The cellulosic binder is particularly better for that formation. Any particular limitation is imposed on the kinds of binder, as long as they exhibit a proper degree of viscosity as a water soluble solvent. However, in the case of the oxidizing agent containing a phase stabilized ammonium nitrate, if it is used in combination with an anionic binder, an ionic reaction is produced to



cause the heat resistance to be significantly reduced, for the reason of which that combination is undesirable. In this case, a nonionic binder should preferably be used. It is preferable that silicon nitride is used as the slag forming agent. The addition of silicon nitride can provide good slag collecting properties without decreasing the combustion speed.

Still another preferable combination of components of the gas generant composition of the present invention is the combination of 5-aminotetrazole and guanidine nitrate used as the fuel, strontium nitrate and ammonium perchlorate used as the oxidizing agent, polyacrylamido used as the binder, and a silane compound used for the purpose of improving moldability at the extrusion molding. This gas generant composition preferably contains 10-30 weight% of 5-aminotetrazole, 5-30 weight% of guanidine nitrate, 10-50 weight% of strontium nitrate, 10-50 weight% of ammonium perchlorate, 0.5-10 weight% of polyacrylamido, and 0.5-10 weight% of silane compound. It is preferable to add water as the solvent in the extrusion molding process. Preferably, silane compound having the property of being soluble in water is then used.

The gas generant composition of the present invention may selectively be formed into a granulated form, a granular form or a pellet form. Also, the mixed conditioner may be formed in the press molding or extrusion molding. It may selectively be formed into a pellet form, a single-hole or a multi-hole (porous) tubular form, for example.

Then, reference will be given to the manufacturing method of the gas generant composition. The gas generant composition of the present

invention can be manufactured in either method of the press molding and the extrusion molding. After having been molded, the gas generant composition is heat-treated to be fully dried out, so as to prevent an ignition lag caused by the moisture and provide an improved environmental  
5 resistance.

When the gas generant composition is molded by the press molding, the anti-caking agent is added to the fuel components and the oxidizing agent, first. Then, the respective components are mixed with a V-type blending machine and then are pulverized. After a prescribed quantity of  
10 pulverized fuel components, pulverized oxidizing agent and auxiliary agent for molding are taken by measurement, they are mixed equally with the V-type blending machine. Thereafter, they are put into the press molding machine and then heat-treated. The gas generating molded products thus obtained are used as the gas generant composition.

15 Likewise, when the gas generant composition is molded by the extrusion molding, the fuel components and the oxidizing agent are pulverized, first. Then, after the respective components are taken by measurement and 8-25 weight% water in outer percentage is added thereto, they are fully kneaded to produce moist agents having viscosity. Thereafter,  
20 they are formed into a desired form and cut properly by use of a vacuum kneading extrusion machine and then are subjected to heat treatment. The gas generating molded products thus obtained are used as the gas generant composition.

The present invention will be further detailed with reference to the

examples.

[Example 1]

24.7 parts by weight of 5-aminotetrazole ( $15\mu\text{m}$  in 50% particle diameter) and 11.9 parts by weight of guanidine nitrate ( $30\mu\text{m}$  in 50% particle diameter) used as the fuel, 53.4 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming agent, and 5.0 parts by weight of synthetic hydrotalcite ( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by use of the V-type blending machine. Then, 15 warts by weight of water to the total quantity of mixed powders was sprayed on the powders under stirring. Thereafter, they were subjected to the wet granulation process to produce granulated powders of not more than 1mm in particle diameter. After the granulated powders thus produced were dried by heating, they were pressed into shapes by a rotary pelletizing machine to produce pellets of 5mm in diameter and 1.5mm in height of the gas generant composition of the present invention.

40g of pellets thus produced were packed in the gas generator 1 shown in FIG. 1. The gas generator 1 comprises a central igniting chamber 7 in which an igniter 2 and a transfer charge 3 are arranged, a combustion chamber 8 which is located around the central igniting chamber and in which gas generants 4 are packed, and a cooling/filtering chamber 9 which is located around the combustion chamber and in which a woven metal wire 5 is disposed. The combustion gas is exhausted out from gas

exhausting holes 6 through the cooling/filtering chamber 9. After having been mounted to a container having an internal volume of 60 liter, the gas generator 1 was operated to discharge the gas into the container, whereby the change of pressure in container with time was measured. The 60 liter tank test results are shown in TABLE 1 of FIG. 3.

[Example 2]

19.7 parts by weight of 5-aminotetrazole ( $15\mu\text{m}$  in 50% particle diameter) and 19.7 parts by weight of guanidine nitrate ( $10\mu\text{m}$  in 50% particle diameter) used as the fuel, 50.6 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming agent, and 5.0 parts by weight of synthetic hydrotalcite ( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by use of the V-type blending machine. Then, 15 weight% of water to the total quantity of mixed powders was sprayed on the powders under stirring. Thereafter, they were subjected to the wet granulation process to produce granulated powders of not more than 1mm in particle diameter. After the granulated powders thus produced were dried by heating, they were pressed into shapes by the rotary pelletizing machine to produce pellets of 5mm in diameter and 1.5mm in height of the gas generant composition of the present invention.

40g of pellets thus produced were packed in the gas generator 1 shown in FIG. 1 and the same test as that of Example 1 was performed. The results obtained are shown in the graph of FIG. 2 and TABLE 1 of FIG.

3. In the graph of FIG. 2, the result of this Example is indicated by a line  
a.

[Example 3]

19.4 parts by weight of 5-aminotetrazole ( $15\mu\text{m}$  in 50% particle  
5 diameter) and 19.4 parts by weight of guanidine nitrate ( $10\mu\text{m}$  in 50%  
particle diameter) used as the fuel, 44.2 parts by weight of strontium nitrate  
( $13\mu\text{m}$  in 50% particle diameter) and 7.0 parts by weight of potassium  
nitrate ( $35\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0  
10 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as  
the slag forming agent, and 5.0 parts by weight of synthetic hydrotalcite  
( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by  
use of the V-type blending machine. Then, 15 weight% of water to the total  
quantity of mixed powders was sprayed on the powders under stirring.  
Thereafter, they were subjected to the wet granulation process to produce  
15 granulated powders of not more than 1mm in particle diameter. After the  
granulated powders thus produced were dried by heating, they were pressed  
into shapes by the rotary pelletizing machine to produce pellets of 6mm in  
diameter and 1.5mm in height of the gas generant composition of the  
present invention.

20 40g of pellets thus produced were packed in the gas generator 1  
shown in FIG. 1 and the same test as that of Example 1 was performed.  
The results obtained are shown in TABLE 1 of FIG. 3.

[Example 4]

41.5 parts by weight of nitroguanidine ( $20\mu\text{m}$  in 50% particle

diameter) and 8.2 parts by weight of guanidine nitrate ( $10\mu\text{m}$  in 50% particle diameter) used as the fuel, 35.3 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) and 5.0 parts by weight of potassium nitrate ( $35\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming agent, and 5.0 parts by weight of synthetic hydrotalcite ( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by use of the V-type blending machine. Then, 15 weight% of water to the total quantity of mixed powders was sprayed on the powders under stirring.

Thereafter, they were subjected to the wet granulation process to produce granulated powders of not more than 1mm in particle diameter. After the granulated powders thus produced were dried by heating, they were pressed into shapes by the rotary pelletizing machine to produce pellets of 5mm in diameter and 2.0mm in height of the gas generant composition of the present invention.

35g of pellets thus produced were packed in the gas generator 1 shown in FIG. 1 and the same test as that of Example 1 was performed. The results obtained are shown in TABLE 1 of FIG. 3.

#### [Example 5]

42.1 parts by weight of nitroguanidine ( $20\mu\text{m}$  in 50% particle diameter) and 8.7 parts by weight of guanidine nitrate ( $30\mu\text{m}$  in 50% particle diameter) used as the fuel, 39.2 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag

forming agent, and 5.0 parts by weight of methyl cellulose used as the binder were dry blended by use of the V-type blending machine. Then, 15 weight% of water to the total quantity of mixed powders was sprayed on the powders under stirring. Thereafter, they were vacuum deaerated while  
5 being kneaded by use of a kneader. The clayey gas generant composition thus obtained was molded into shapes by use of a screw extrusion machine and then was dried by heating to thereby produce columnar molded products of gas generant composition of 3mm in diameter and 2mm in height.

10 35g of molded products thus produced were packed in the gas generator 1 shown in FIG. 1 and the same test as that of Example 1 was performed. The results obtained are shown in the graph of FIG. 2 and TABLE 1 of FIG. 3. In the graph of FIG. 2, the result of this Example is indicated by a line b.

15 [Example 6]

34.7 parts by weight of nitroguanidine ( $20\mu\text{m}$  in 50% particle diameter) and 9.5 parts by weight of guanidine nitrate ( $10\mu\text{m}$  in 50% particle diameter) used as the fuel, 36.8 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) and 10.5 parts by weight of basic copper  
20 nitrate ( $11\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 3.5 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming agent, and 5.0 parts by weight of methyl cellulose used as the binder were dry blended by use of the V-type blending machine. Then, 15 weight% of water to the total quantity of mixed powders was sprayed on

the powders under stirring. Thereafter, they were vacuum deaerated while being kneaded by use of the kneader. The clayey gas generant composition thus obtained was molded into shapes by use of the screw extrusion machine and then was dried by heating to thereby produce columnar  
5 molded products of gas generant composition of 4mm in diameter and 2mm in height.

35g of molded products thus produced were packed in the gas generator 1 shown in FIG. 1 and the same test as that of Example 1 was performed. The results obtained are shown in TABLE 1 of FIG. 3.

10 [Comparative Example 1]

30.9 parts by weight of 5-aminotetrazole ( $15\mu\text{m}$  in 50% particle diameter) used as the fuel, 57.9 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming  
15 agent, and 5.0 parts by weight of synthetic hydrotalcite ( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by use of the V-type blending machine. Then, 15 parts by weight of water to the total quantity of mixed powders was sprayed on the powders under stirring. Thereafter, they were subjected to the wet granulation process to produce granulated  
20 powders of not more than 1mm in particle diameter. After the granulated powders thus produced were dried by heating, they were pressed into shapes by the rotary pelletizing machine to produce pellets of gas generant composition of 5mm in diameter and 2.0mm in height.

40g of pellets thus produced were packed in the gas generator 1



shown in FIG. 1 and the same test as that of Example 1 was performed.

The results obtained are shown in the graph of FIG. 2 and TABLE 1 of FIG.

3. In the graph of FIG. 2, the result of this Comparative Example is indicated by a line c.

#### 5 [Comparative Example 2]

50.6 parts by weight of guanidine nitrate ( $15\mu\text{m}$  in 50% particle diameter) used as the fuel, 39.4 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming agent, and 5.0 parts by weight of synthetic hydrotalcite ( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by use of the V-type blending machine. Then, 15 parts by weight of water to the total quantity of mixed powders was sprayed on the powders under stirring. Thereafter, they were subjected to the wet granulation process to produce granulated powders of not more than 1mm in particle diameter. After the granulated powders thus produced were dried by heating, they were pressed into shapes by the rotary pelletizing machine to produce pellets of gas generant composition of 5mm in diameter and 1.5mm in height.

40g of pellets thus produced were packed in the gas generator 1

shown in FIG. 1 and the same test as that of Example 1 was performed.

The results obtained are shown in the graph of FIG. 2 and TABLE 1 of FIG.

3. In the graph of FIG. 2, the result of this Comparative Example is indicated by a line d.

#### [Comparative Example 3]

24.7 parts by weight of 5-aminotetrazole ( $15\mu\text{m}$  in 50% particle diameter) and 11.9 parts by weight of guanidine nitrate ( $50\mu\text{m}$  in 50% particle diameter) used as the fuel, 53.4 parts by weight of strontium nitrate ( $13\mu\text{m}$  in 50% particle diameter) used as the oxidizing agent, 5.0 parts by weight of silicon nitride ( $5\mu\text{m}$  in 50% particle diameter) used as the slag forming agent, and 5.0 parts by weight of synthetic hydrotalcite ( $10\mu\text{m}$  in 50% particle diameter) used as the binder were dry blended by use of the V-type blending machine. Then, 15 parts by weight of water to the total quantity of mixed powders was sprayed on the powders under stirring.

Thereafter, they were subjected to the wet granulation process to produce granulated powders of not more than 1mm in particle diameter. After the granulated powders thus produced were dried by heating, they were pressed into shapes by the rotary pelletizing machine to produce pellets of gas generant composition of 5mm in diameter and 1.5mm in height.

40g of pellets thus produced were packed in the gas generator 1 shown in FIG. 1 and the same test as that of Example 1 was performed. The results obtained are shown in the graph of FIG. 2 and TABLE 1 of FIG. 3. In the graph of FIG. 2, the result of this Comparative Example is indicated by a line e.

As clearly seen from the results of FIGS. 2 and 3, in the 60 liter tank test, the molded products of the gas generant composition of Examples 1-6 of the present invention provide the results that the gas generating speed increases in a controlled rate in the period of 10-20ms from the ignition and then increases sharply on and after 20ms and the pressure becomes

substantially the same level as that of the conventional gas generant composition on and after 40ms. It is understood from this that the molded products of the gas generant composition of the present invention provided suitable burning performances for the gas generator.

5 In comparison with this, the gas generant composition of Comparative Example 1, in which guanidine nitrate is not added as the fuel, in other words, the fuel consists of only a high-energy nitrogen-containing organic compound, provides the result that the 60 liter tank pressure increases largely in the period of 20ms from the ignition, as seen from FIGS. 2 and 3,  
10 and thus provides an increased harm for the inflator. The gas generant composition of Comparative Example 2, in which the fuel consists of only a low-energy guanidine nitrate, provides excessive reduction in combustion speed, as seen from FIG. 2, and thus is improper as the gas generant. The gas generant composition of Comparative Example 3 provides the result  
15 that when the 50% average particle diameter of guanidine nitrate exceeds  $40\mu\text{m}$ , the 60 liter tank pressure becomes little more than that of Comparative Example 1 and does not provide the effects of the present invention.

As clearly seen from the description above, in the gas generant  
20 composition of the present invention, the fuel comprises at least two nitrogen-containing organic compounds comprising a high-energy nitrogen-containing organic compound exhibiting a high combustion speed and a low-energy nitrogen-containing organic compound exhibiting a low combustion speed, and the low-energy nitrogen-containing organic

compound is made to have the 50% average particle diameter of  $40\mu\text{m}$  or less, whereby the gas generant composition exhibiting a suitable burning behavior for the gas generator is provided. Thus, the gas generator having the combustion performance of doing little harm to the risers can be realized with ease and at low cost by using the gas generant comprising the gas generant composition of the present invention.

#### Capabilities of Exploitation in Industry

The present invention provides an optimum gas generant composition useful for gas generants used for a gas generator for an automobile occupant restraint system, such as an airbag and a pretensioner, which exhibits the burning characteristics to produce a further ideal occupant protecting performance, without any complex production process being required and without any limitation being imposed on the forms of the gas generants.

## Claims

1. A gas generant composition comprising a fuel, an oxidizing agent and an additive, wherein the fuel comprises at least one high-energy  
5 nitrogen-containing organic compound and at least one low-energy nitrogen-containing organic compound, and the low-energy nitrogen-containing organic compound has a 50% average particle diameter of  $40\text{ }\mu\text{m}$  or less.
2. The gas generant composition according to Claim 1, wherein the  
10 low-energy nitrogen-containing organic compound has the 50% average particle diameter of  $20\text{ }\mu\text{m}$  or less.
3. The gas generant composition according to Claim 1, wherein the high-energy nitrogen-containing organic compound comprises at least one material selected from the group consisting of aminotetrazole,  
15 nitroguanidine, and triaminoguanidine nitrate
4. The gas generant composition according to Claim 1, wherein the low-energy nitrogen-containing organic compound is guanidine nitrate.
5. The gas generant composition according to Claim 1, wherein the high-energy nitrogen-containing organic compound comprises at least one  
20 material selected from the group consisting of aminotetrazole, nitroguanidine, and triaminoguanidine nitrate, and the low-energy nitrogen-containing organic compound is guanidine nitrate.
6. The gas generant composition according to Claim 1, wherein the oxidizing agent comprises at least one material selected from the group

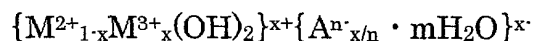
consisting of nitrate, perchlorate, chlorate as concerns alkali metals or alkali earth metals and basic copper nitrate.

7. The gas generant composition according to Claim 1, wherein the oxidizing agent comprises at least one material selected from the group

5 consisting of mixtures of phase stabilized ammonium nitrate or ammonium perchlorate and nitrate, perchlorate, chlorate as concerns alkali metals or alkali earth metals and basic copper nitrate.

8. The gas generant composition according to Claim 1, wherein the additive comprises silicon nitride or silicon carbide.

10 9. The gas generant composition according to Claim 1, wherein the additive comprises hydrotalcites expressed by the following general formula:



where  $M^{2+}$  represents bivalent metals including  $Mg^{2+}$ ,  $Mn^{2+}$ ,  $Fe^{2+}$ ,  $Co^{2+}$ ,  $Ni^{2+}$ ,  $Cu^{2+}$  and  $Zn^{2+}$ ;

15  $M^{3+}$  represents trivalent metals including  $Al^{3+}$ ,  $Fe^{3+}$ ,  $Cr^{3+}$ ,  $Co^{3+}$  and  $In^{3+}$ ;

$A^{n-}$  represents n-valence anions including  $OH^-$ ,  $F^-$ ,  $Cl^-$ ,  $NO_3^-$ ,  $CO_3^{2-}$ ,  $SO_4^{2-}$ ,  $Fe(CN)_6^{3-}$ ,  $CH_3COO^-$ , ion oxalate, and ion salicylate; and

X:  $0 < x \leq 0.33$ .

20 10. The gas generant composition according to Claim 9, wherein the hydrotalcites comprise synthetic hydrotalcite expressed by the chemical formula of  $Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$  or pyroaurite expressed by the chemical formula of  $Mg_6Fe_2(OH)_{16}CO_3 \cdot 4H_2O$ .

11. The gas generant composition according to Claim 1, wherein the

additive comprises at least one cellulosic binders selected from the group consisting of carboxymethyl cellulose, methyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, and hydroxylpropyl methyl cellulose or natural polymer.

5 12. The gas generant composition according to Claim 1, wherein the additive comprises at least one material selected from the group consisting of polyacrylic acid, sodium polyacrylate, polyacrylamide, and two or three copolymerized compounds thereof.

10 13. The gas generant composition according to Claim 1, wherein the additive is a silane compound.

14. The gas generant composition according to Claim 1, wherein the fuel consists of 5-aminotetrazole and guanidine nitrate, the oxidizing agent consists of strontium nitrate, and the additive consists of silicon nitride and synthetic hydrotalcite.

15 15. The gas generant composition according to Claim 1, wherein the fuel consists of 5-aminotetrazole and guanidine nitrate, the oxidizing agent consists of strontium nitrate and potassium nitrate, and the additive consists of silicon nitride and synthetic hydrotalcite.

20 16. The gas generant composition according to Claim 1, wherein the fuel consists of 5-aminotetrazole and guanidine nitrate, the oxidizing agent consists of strontium nitrate and basic copper nitrate, and the additive consists of silicon nitride and synthetic hydrotalcite.

17. The gas generant composition according to Claim 1, wherein the fuel consists of nitroguanidine and guanidine nitrate, the oxidizing agent

consists of strontium nitrate, and the additive consists of silicon nitride.

18. The gas generant composition according to Claim 1, wherein the fuel consists of nitroguanidine and guanidine nitrate, the oxidizing agent consists of strontium nitrate and potassium nitrate, and the additive  
5 consists of silicon nitride.

19. The gas generant composition according to Claim 1, wherein the fuel consists of nitroguanidine and guanidine nitrate and the oxidizing agent consists of strontium nitrate and basic copper nitrate.

20. The gas generant composition according to Claim 1, wherein the fuel  
10 consists of 5-aminotetrazole and guanidine nitrate, the oxidizing agent consists of strontium nitrate and ammonium perchlorate, and the additive consists of polyacrylamide and silane compound.

21. The gas generant composition according to Claim 1, which comprises  
15 10-30 weight% of 5-aminotetrazole as the high-energy nitrogen-containing organic compound, 5-30 weight% of guanidine nitrate as the low-energy nitrogen-containing organic compound, 30-70 weight% of strontium nitrate as the oxidizing agent, and 0.5-10 weight% of silicon nitride and 2-10 weight% of synthetic hydrotalcite as the additive.

22. The gas generant composition according to Claim 1, which comprises  
20 20-55 weight% of nitroguanidine as the high-energy nitrogen-containing organic compound, 5-30 weight% of guanidine nitrate as the low-energy nitrogen-containing organic compound, 30-60 weight% of strontium nitrate as the oxidizing agent, and 0.5-10 weight% of silicon nitride and 2-10 weight% of synthetic hydrotalcite as the additive.



23. The gas generant composition according to Claim 1, which comprises 20-55 weight% of nitroguanidine as the high-energy nitrogen-containing organic compound, 5-30 weight% of guanidine nitrate as the low-energy nitrogen-containing organic compound, 30-60 weight% of strontium nitrate as the oxidizing agent, and 0.5-10 weight% of silicon nitride and 2-10 weight% of cellulosic binder as the additive.

24. The gas generant composition according to Claim 1, which comprises 10-30 weight% of 5-aminotetrazole as the high-energy nitrogen-containing organic compound, 5-30 weight% of guanidine nitrate as the low-energy nitrogen-containing organic compound, 10-50 weight% of strontium nitrate and 10-50 weight% of ammonium perchlorate as the oxidizing agent, and 0.5-10 weight% of polyacrylamide and 0.5-10 weight% of silane compound as the additive.

25. The gas generant composition according to any one of Claims 21 through 24, which further comprises not more than 10 weight% of potassium nitrate.

26. The gas generant composition according to any one of Claims 21 through 24, which further comprises not more than 30 weight% of basic copper nitrate.

27. The gas generant composition according to Claim 23, wherein the cellulosic binder comprises at least one material selected from the group consisting of carboxymethyl cellulose, methyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, and hydroxylpropyl methyl cellulose.

FIG. 1

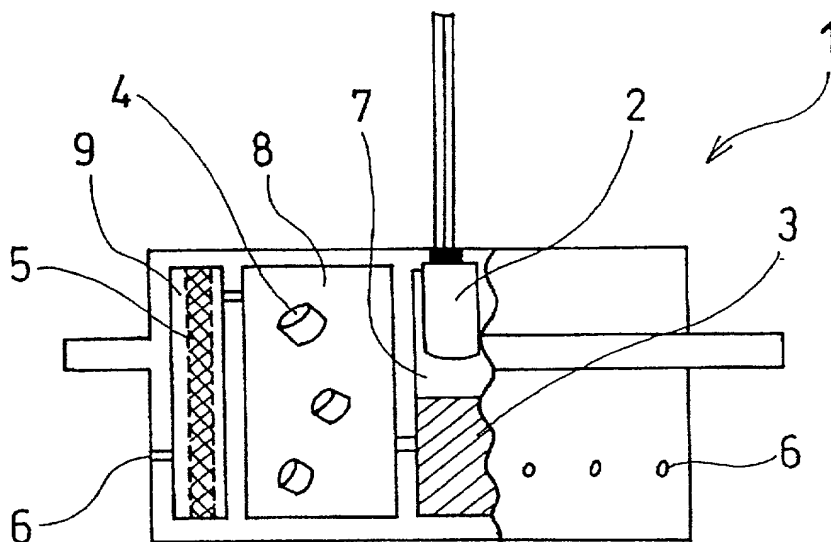


FIG. 2

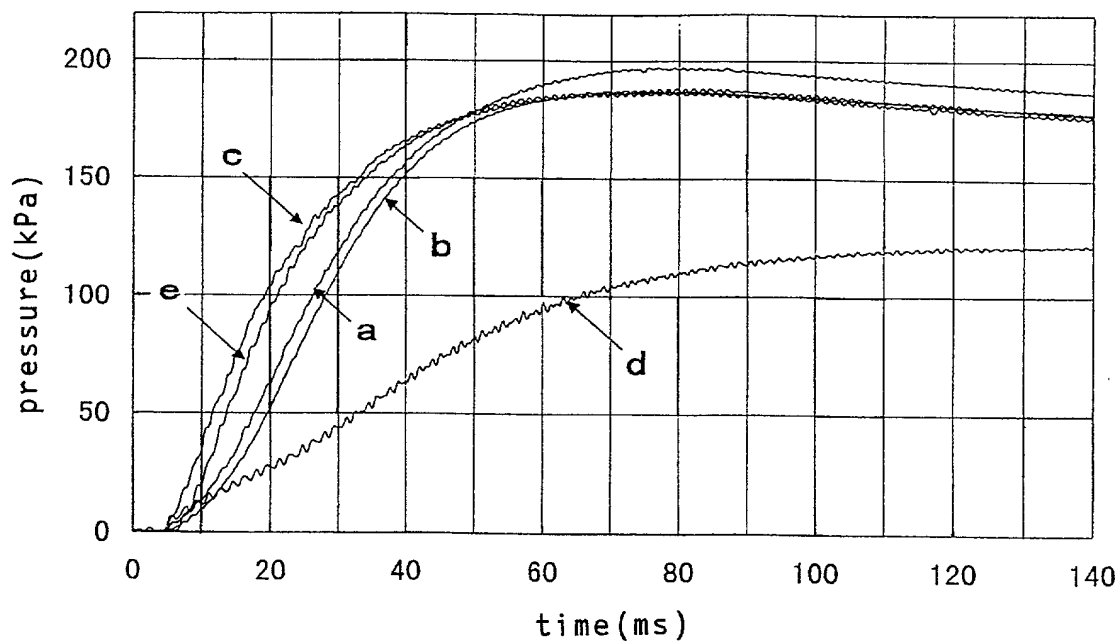


FIG. 3

TABLE 1 60 liter tank test pressure (kPa)

	10ms	20ms	30ms	40ms	50ms
Example 1	18.0	62.0	136.0	158.0	178.0
Example 2	17.0	60.0	136.5	163.0	182.0
Example 3	17.5	63.0	138.5	166.0	178.0
Example 4	15.0	69.0	141.0	169.5	180.0
Example 5	16.0	53.0	119.0	151.0	174.0
Example 6	16.5	57.5	124.0	160.0	175.0
Comparative Example 1	37.0	105.0	143.0	166.0	177.0
Comparative Example 2	12.0	27.0	44.0	63.0	82.0
Comparative Example 3	24.0	97.0	138.0	163.0	178.0

Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者（下記の名称が複数の場合）であると信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled.

Gas Generant Composition

上記発明の明細書は、

☐ 本書に添付されています。

the specification of which

☐ is attached hereto.

☒ 月 日に提出され、米国出願番号または特許協力条約国際出願番号を \_\_\_\_\_ とし、  
(該当する場合) \_\_\_\_\_ に訂正されました。

☒ was filed on June 22, 2000

as United States Application Number or

PCT International Application Number

PCT/JP00/04070 and was amended on

\_\_\_\_\_ (if applicable).

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

## Japanese Language Declaration

(日本語宣言書)

私は、米国法典第35編119条(a)-(d)項又は365条(b)項に基づき下記の、米国以外の国の少なくとも一方国を指定している特許協力条約365(a)項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

## Prior Foreign Application(s)

外国での先行出願

11-180178	Japan
(Number)	(Country)
(番号)	(国名)
11-298989	Japan
(Number)	(Country)
(番号)	(国名)
(Number)	(Country)
(番号)	(国名)
(Number)	(Country)
(番号)	(国名)

私は、第35編米国法典119条(e)項に基づいて下記の米国特許出願規定に記載された権利をここに主張いたします。

(Application No.)	(Filing Date)
(出願番号)	(出願日)

私は、下記の米国法典第35編120条に基づいて下記の米国特許出願に記載された権利、又は米国を指定している特許協力条約365条(c)に基づく権利をここに主張します。また、本出願の各請求範囲の内容が米国法典第35編112条第1項又は特許協力条約で規定された方法で先行する米国特許出願に開示されていない限り、その先行米国出願書提出日以降で本出願書の日本国内または特許協力条約国際提出日までの期間中に入手された、連邦規則法典第37編1条56項で定義された特許資格の有無に関する重要な情報について開示義務があることを認識しています。

PCT/JP00/04070	June 22, 2000
(Application No.)	(Filing Date)
(出願番号)	(出願日)

(Application No.)	(Filing Date)
(出願番号)	(出願日)

私は、私自身の知識に基づいて本宣言書中で私が行なう表明が真実であり、かつ私の入手した情報と私が信じているところに基づく表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第18編第1001条に基づき、罰金または拘禁、もしくはその両方により処罰されること、そしてそのような故意による虚偽の声明を行なえば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに上記のごとく宣誓を致します。

I hereby claim foreign priority under Title 35, United States code, Section119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or Section365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

## Priority Claimed

優先権主張

25/06/1999	<input checked="" type="checkbox"/>	<input type="checkbox"/>
(Day/Month/Year Filed)	Yes	No
(出願年月日)	はい	いいえ
21/10/1999	<input checked="" type="checkbox"/>	<input type="checkbox"/>
(Day/Month/Year Filed)	Yes	No
(出願年月日)	はい	いいえ
(Day/Month/Year Filed)	<input type="checkbox"/>	<input type="checkbox"/>
(出願年月日)	Yes	No
(Day/Month/Year Filed)	はい	いいえ
(出願年月日)	<input type="checkbox"/>	<input type="checkbox"/>
(出願年月日)	Yes	No
(出願年月日)	はい	いいえ

I hereby claim the benefit under Title 35, United States Code, Section 119 (e) of any United States provisional application(s) listed below.

(Application No.)	(Filing Date)
(出願番号)	(出願日)

I hereby claim the benefit under Title35, United States Code, Section 120 of any United States application(s), or Section365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title35, United States Code Section112, I acknowledge the the duty to disclose information which is material to patentability as defined in Title37, Code of Federal Regulations, Section1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

Pending

(Status: Patented, Pending, Abandoned)
(現況: 特許許可済、係属中、放棄済)

(Status: Patented, Pending, Abandoned)
(現況: 特許許可済、係属中、放棄済)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

## Japanese Language Declaration

(日本語宣言書)

委任状：私は下記の発明者として、本出願に関する一切の手続きを米特許商標局に対して遂行する弁理士または代理人として、下記の者を指名いたします。

(弁理士、または代理人の指名及び登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: (list name and registration number)

**022850**

書類送付先

Send Correspondence to:

**022850**

直接電話連絡先：(名前及び電話番号)

Direct Telephone Calls to: (name and telephone number)  
(703) 413-3000

唯一または第一発明者名

Full name of sole or first inventor

Dairi KUBO

発明者の署名

日付

Inventor's signature

Date

Dairi Kubo

OCT. 4. 2001

住所

Residence

3-3-14, Kitahirano, Himeji-shi,  
Hyogo 670-0893 Japan

国籍

Citizenship

JAPAN

郵便の宛先

Post Office Address

same as above

第二共同発明者

Full name of second joint inventor, if any

Eishi SATO

第二共同発明者の署名

日付

Second Inventor's signature

Date

Eishi Sato

OCT. 4. 2001

住所

Residence

3903-39, Toyotomi, Toyotomi-cho, Himeji-shi,  
Hyogo 679-2123 Japan

国籍

Citizenship

JAPAN

郵便の宛先

Post Office Address

same as above

(第三以降の共同発明者についても、同様に記載し、署名すること)

(Supply similar information and signature for third and subsequent joint inventors.)

Page 3 of 4

## Japanese Language Declaration

第三共同発明者	Full name of third joint inventor, if any Ryo i KODAMA	
第三共同発明者の署名	Third Inventor's signature Ryoi Kodama	Date OCT. 4. 2001
住所	Residence 3903-39, Toyotomi, Toyotomi-cho, Himeji-shi, Hyogo 679-2123 Japan JPX	
国籍	Citizenship JAPAN	
郵便の宛先	Post Office Address same as above	
第四共同発明者	Full name of fourth joint inventor, if any Kenjiro IKEDA	
第四共同発明者の署名	Fourth Inventor's signature Kenjiro Ikeda	Date OCT. 4. 2001
住所	Residence 746-3, Mikage, Toyotomi-cho, Himeji-shi, Hyogo 679-2122 Japan JPX	
国籍	Citizenship JAPAN	
郵便の宛先	Post Office Address same as above	
第五共同発明者	Full name of fifth joint inventor, if any	
第五共同発明者の署名	Fifth Inventor's signature	Date
住所	Residence	
国籍	Citizenship	
郵便の宛先	Post Office Address	
第六共同発明者	Full name of sixth joint inventor, if any	
第六共同発明者の署名	Sixth Inventor's signature	Date
住所	Residence	
国籍	Citizenship	
郵便の宛先	Post Office Address	

(第七またはそれ以降の共同発明者に対しても同様な情報および署名を提供すること。)

(Supply similar information and signature for seventh and subsequent joint inventors.)